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"High Power Broadly Tunable Picosecond IR Laser System for Use in Nonlinear Spectroscopic Applications"

by

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Abstract

A picosecond laser system which will generate high power tunable IR pulses with bandwidths suitable for spectroscopic applications is discussed. The system is based on white light continuum generation in ethylene glycol and optical parametric amplification in potassium titanyl phosphate. The non-linear optical processes are driven by a regeneratively amplified Ti:sapphire laser which produces 1.7 ps pulses at a repetition rate of 1 kHz. Energies as high as 40 μJ and 12 μJ have been achieved over the signal, 1.02-1.16 μm, and idler, 2.6-3.7 μm, tuning ranges respectively. The IR beam temporal and spatial characteristics are also presented.

Nonlinear optical methods such as second harmonic generation (SHG) and sum frequency generation (SFG) are uniquely suited to the study of buried interfaces. In the case of visible/IR sum frequency generation one can achieve both molecular and interfacial selectivity. Because of this unique selectivity the use of SFG as a spectroscopic tool to study interfaces at a molecular level has grown rapidly over the last several years[1,2,3]. One difficulty encountered in SFG studies of molecular systems has been the lack of short pulse, high power tunable infrared laser systems which can be used to probe the vibrational modes of condensed phase interfacial molecular species. Since the ability to resolve closely spaced vibrations directly depends on the spectral extent of the IR pulses, the bandwidth of the pulses also becomes a major consideration. Ti:sapphire systems show promise for nonlinear spectroscopic experiments which benefit from the short pulses, and thus high peak powers, that these lasers provide. However, for the majority of cases the short pulses do not have the narrow bandwidth necessary for spectral resolution of vibrational modes. In this letter we describe a Ti:sapphire based laser system which is optimized for providing both high peak powers and relatively narrow bandwidths. It consists of a Ti:sapphire regenerative amplifier which we use to generate high power picosecond IR laser pulses tunable from 1 μm to 3.8 μm.

Parametric processes such as optical parametric generation (OPG) and amplification (OPA) in nonlinear crystals have been used extensively to generate tunable IR pulses[4,5]. The majority of these systems are based on Nd:YAG pump lasers and produce pulses with durations from tens of picoseconds to nanoseconds[6,7,8] and bandwidths from several wavenumbers to a few tenths of a wavenumber. More recently regeneratively amplified Ti:sapphire lasers have been used to produce subpicosecond tunable IR pulses[9,10,11] with bandwidths on the order of hunderds of wavenumbers. We employ a Ti:sapphire regenerative amplifier which produces 1.7 picosecond pulses at 800 nm and a bandwidth of 21 cm^{-1} as our pump source. The IR generation is based on white light continuum generation in ethylene glycol and optical parametric amplification (OPA) in two

independently pumped potassium titanyl phosphate (KTP) crystals. White light generation was chosen to provide an intense seed covering the entire signal wavelength region, 1-1.6 μm. Our first attempt at IR generation involved seeding the first KTP crystal with the entire IR portion of the continuum from 1 μm to 1.6 μm. This, however, produced signal and idler bandwidths in excess of 100 cm^{-1}, the large bandwidth being a result of the acceptance bandwidth of KTP which is quite large in the 2.6-3.8 μm region. The bandwidth can be narrowed by supplying the KTP crystal with a narrower seed. This was accomplished by using a grating to spatially disperse the continuum resulting in a seed bandwitdth of approximately 9 cm^{-1}. KTP was selected as the nonlinear crystal because of the desired tuning range, 1-4 μm, its figure of merit which is proportional to the conversion efficiency, and its availability. Further, type II (ordinary idler, extraordinary signal, ordinary pump) phase matching in the *xz* plane of the KTP crystals was chosen because of a higher[12] d_{eff} and larger tuning range for an 800 nm pump. We use two amplification stages mainly because higher energies can be achieved but also because a second crystal angle tuned in the opposite direction has the effect of compensating for the beam displacement from the first crystal.

The Ti:sapphire regenerative amplifier laser system consists of a Ti:sapphire oscillator (Coherent Mira-900 basic) pumped by an Ar^+ ion laser (Coherent Innova-310), a stretcher-compressor (Quantronix 4822), and a Ti:sapphire regenerative amplifier (Quantronix 4810) pumped by the second harmonic of a Nd:YLF laser (Quantronix 527 DP-H). The output of the laser system (750 μJ, 800nm, and 1 kHz repetition rate) is first split with an 80% reflective mirror which sends 600 μJ to the optics for nonlinear conversion. Figure 1 depicts the optical set-up used to convert the 800 nm photons into IR photons. The 600 μJ is split into three pump lines with two 50% reflecting mirrors, BS1 and BS2. The three lines are used for pumping the white light generator WL and the two KTP crystals OPA1 and OPA2. The white light pump passes through lens L1 which focuses 125 μJ of the 800 nm light into the ethylene glycol. The ethylene glycol flows

through a 10x10x50 mm quartz cell (WL) which limits the effect of bubble formation on the white light generated caused by boiling. The effect of bubble formation is also minimized by cooling the ethylene glycol to approximately 16 degrees. The white light generated then passes through filter F1 (RG-1000) which transmits only the IR portion of the continuum. The IR portion of the continuum is brought to a focus at the position of OPA1 by lens L2. The bandwidth of the seed supplied OPA1 is narrowed by grating G1 (600 g/mm, blazed at 750 nm) located 400 mm from OPA1. The white light seed is then amplified in OPA1 with 145 μJ of the 800 nm light focused to a beam diameter, 2ω, of 440 μm by lenses L4 and L5. The IR seed and the 800 nm pump are combined with dichroic mirror M1 and the temporal overlap of the seed and the pump in OPA1 is controlled by delay line D1. After this first stage of amplification signal energies as high as 20 μJ and idler energies as high as 4 μJ are obtained. Since higher energies are desired another amplification stage (OPA2) located 560 mm from OPA1 is employed. Filter F2 (75% transmission from 1.7-3.4 μm) is used to transmit the idler for amplification in OPA2. The idler is used for the seed in OPA2 because higher energies and better spatial beam profiles are attained in this configuration (see following discussion). The idler from OPA1 is focused to a beam diameter, 2ω, of 760 μm at the position of OPA2 by a CaF_2 lens, L1. The idler generated in OPA1 is amplified in OPA2 with 200 μJ of the 800 nm light which is focused to a beam diameter, 2ω, of 600 μm by lenses L6 and L7. The idler and the 800 nm pump are combined with the use of dichroic mirror M2 and the temporal overlap of the seed and the pump in OPA2 is controlled by a second delay line D2. One of two filters is used in the position of F3; the first for the measurement of the idler energy (2.5 μm longpass) and the other for the measurement of the signal energy (RG-1000).

The output of the laser system was characterized in terms of wavelength, power, bandwidth, pulse duration, and spatial beam quality. Wavelengths were measured with a 0.3 meter crossed Czerny-Turner monochromator with a 300 groove/mm grating and a

indium antimonide detector. Bandwidths (full width at half maximum intensity, FWHM) were measured with the monochromator or with a grating (1200 or 1800 groove/mm) in combination with a one dimensional CCD array (UniData BP2048). The resolution using the monochromator was approximately 1 cm^{-1}, while the resolution of the grating/CCD combination was approximately 4 cm^{-1} and depended on the diameter of the beam used. Powers were measured with a pyroelectric joule meter, and pulse durations characterized by autocorrelation (Inrad 5-14B) and/or cross correlation widths (FWHM). Spatial beam quality was characterized using knife-edge measurements across the horizontal transverse profile of the beams. Assuming an approximately gaussian transverse spatial profile, the distance between knife-edge positions passing 16% and 84% of the total beam energy was used to approximate the gaussian beam radius ω, which has the standard definition of the half width at $1/e$ of maximum for the field *amplitude*, or the half width at $1/e^2$ of maximum for the *intensity*[13].This is in turn related to the FWHM for the intensity by ω = FWHM/$(2\ln 2)^{1/2}$. The measured pump and idler beam sizes were then used to estimate the M^2 parameter for the idler beam[14].

Theoretical tuning curves were generated by imposing Type II (*oeo*) collinear phase matching in the *xz* plane (ϕ=0°) of the KTP crystal[15] . The Sellmeier constants used are from Vanherzeele[12]. Both the theoretical and experimental tuning curves over a practical tuning range are shown in Figure 2. A practical tuning range is limited by relative misalignment of the pump and seed beams in the KTP crystals which occurs as they are rotated. With intermediate realignment, the actual tuning curve extends from beyond 3.8 μm to 1.6 μm (the degeneracy point for an 800 nm pump wavelength) for the idler and from 1.6 μm to 1.01 μm for the signal.

The tuning curve in Figure 2 was generated by angle tuning both the KTP crystals and the grating. As was mentioned earlier the bandwidth of the output IR pulses when seeded with the entire IR portion of the continuum is greater than 100 cm^{-1}. By employing a diffraction

grating so that the first order diffracted beam is propagated through the optical parametric amplifier, the bandwidth of the amplified signal beam was reduced to about 31 cm^{-1}. A rough calculation of the bandwidth expected to fall within the spatial profile of the pump beam in the first KTP crystal (260 μm FWHM) yields $\Delta\bar{\nu} \approx a\,\bar{\nu}^2 \cos\beta\,(d/D) = 8.6\,cm^{-1}$ where *a* is the groove spacing of the grating, *d* is the diameter of the pump beam (FWHM), *D* is the distance from the grating to the first KTP crystal, and β is angle of diffraction from the grating. This shows that by narrowing the seed bandwidth, the bandwidth of the amplified signal should be limited by the pump bandwidth (21 cm^{-1}) and that the bandwidth achieved here is somewhat larger than this lower limit. A bandwidth of 27 cm^{-1} was measured for the idler using the monochromator. Bandwidths of 25-32 cm^{-1} were obtained through out the tuning range shown in Figure 2 for both signal and idler pulses.

The output from the first stage of parametric amplification consists of an amplified signal pulse and the corresponding idler pulse. Due to group velocity mismatch effects, these pulses are not well overlapped temporally upon exiting the first KTP crystal, so only one of these two pulses can be effectively amplified in the second KTP crystal. The idler was chosen for several reasons. First, the wavelength region of interest to us experimentally corresponds to the idler wavelengths, and by seeding the second crystal with the idler up to 50% more energy can be obtained in the idler pulses than by seeding with the signal. Second, it was observed that the output beam profile from the second KTP crystal was qualitatively better when seeded with the idler. When seeded with the signal, the output of the second parametric amplifier stage was more divergent and had an elliptical transverse profile, possibly due to acceptance angle or acceptance bandwidth effects in the KTP crystal. This was not investigated further.

The propagation of the idler beam produced in the first KTP crystal was characterized by a series of knife-edge measurements. Assuming an idler beam waist of ω_0=220 μm (the pump beam size) in the first KTP crystal yielded an estimate of $M^2 \approx 2.5$.

This beam was then focused by lens L1 (see Figure 1) to a spotsize of ω=380 μm in the second KTP crystal. The pump beam in the second crystal had a spotsize of ω= 300 μm. Assuming this value for the idler waist within the second crystal yielded an estimate of $M^2 \approx 2.0$ for the idler beam following the second stage of amplification.

A power curve was generated by rotating the grating and the crystals together with no intervening realignment and is shown in Figure 3. The idler pulse energies were above 5 μJ over a range from 2.7 to 3.5 μm, and over 10 μJ from 2.7 to 3.2 μm and have a relative instability of 4% (standard deviation). It should be emphasized that these curves were generated by aligning the system once at the beginning and making no further adjustments as the wavelength was scanned by rotating the grating and KTP crystals and grating. Another consideration is that the particular filters used in this configuration are not suitable for the entire tuning range. In particular, filter F2 has a nominal transmission range of 1.7-3.4 μm, and so begins to limit the amount of idler available to seed the second KTP crystal as the system is tuned beyond 3.5 μm. Filter F3 has a nominal transmission cutoff below 2.5 μm, limiting the amount of idler energy measurable below that wavelength.

The temporal and bandwidth characteristics of the pump, signal, and idler were measured. It is important to keep in mind the method by which the pump pulses are generated in the Ti:sapphire regenerative amplifier. The grating pulse stretcher employed by the amplifier system contains a spectral filter to reduce the bandwidth, thereby increasing the pulsewidths. The spectral filter results in an essentially square frequency spectrum for the amplified pulses used to pump the continuum generator and parametric amplifiers. A square spectrum corresponds to pulses with an intensity profile of $[(\sin t)/t]^2$, which yields a time/bandwidth product of 0.886[16] and an autocorrelation width to pulse width ratio of 1.33. The output of the Ti:sapphire laser system has a bandwidth of 21 cm^{-1} and a pulse width of 1.7 ps (based on an autocorrelation of 2.2 ps FWHM), resulting in a

time/bandwidth product of 1.05, or 1.2 times transform limited. These pulses were cross correlated with both signal and idler pulses by sum frequency generation in another KTP crystal, yielding cross correlation widths (FWHM) of 3.0 ps and 2.6 ps for the signal and idler pulses, respectively. Pulse widths for the signal and idler were extracted according to the relation $\Delta t_{cc}^2 = \Delta t_p^2 + \Delta t_{s,i}^2$. This yields a pulse width of 2.5 ps and a time bandwidth product of 2.3 for the signal, and 2.0 ps and 1.6 for the idler. The signal and idler spectral profiles did not appear to be square, and no further assumption as to the functional forms for the spectra or temporal pulse profiles have been made.

The intense IR pulses that are produced by this system make it an attractive source for nonlinear spectroscopic applications. In addition the moderately narrow band widths and broad tuning range will allow for the study of a large number of condensed phase interfacial molecules via their vibrational modes. The 25-30 cm^{-1} bandwidths reported here are well suited for studies of this type where the bandwidths of the molecular vibrations are relatively large. However, if still narrower bandwidths are desired the pump laser source can be modified to give slightly longer pulses which will result in a smaller bandwidth. The practical limit of the bandwidths attainable by this type of modification is approximately 10-15 cm^{-1}. The use of other nonlinear crystals such as $AgGaS_2$ to extend the tuning range beyond 3.8 μm would also allow us to study a wider variety of molecules.

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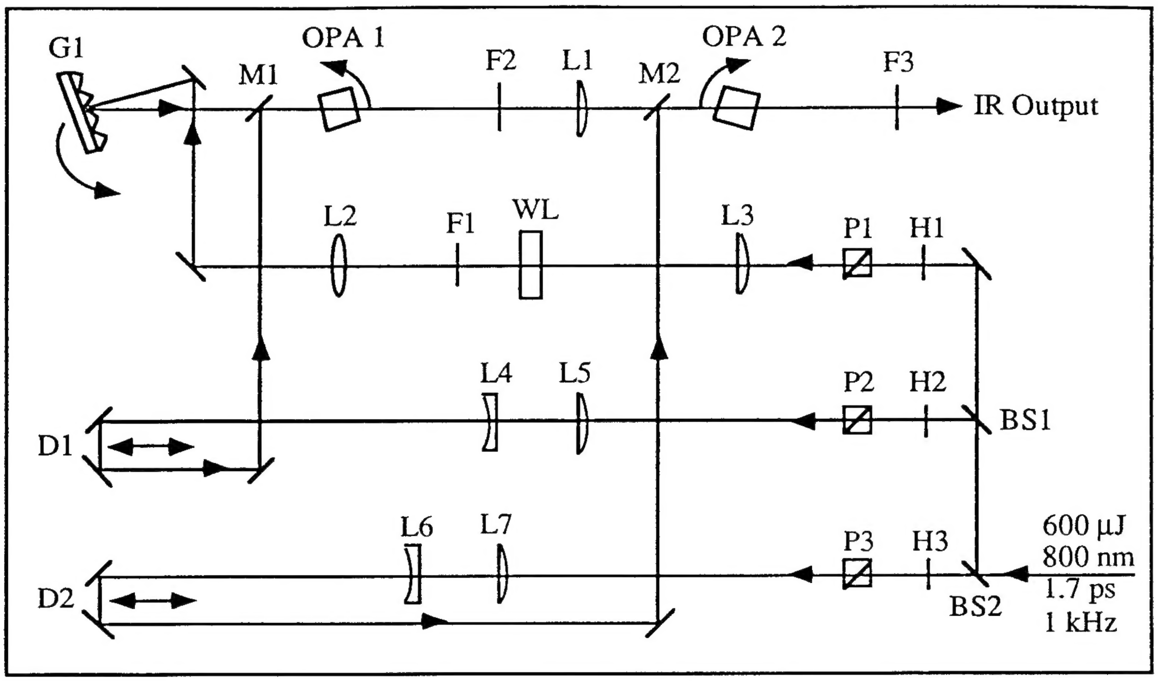

Figure 1

Diagram for OPA system. D1 and 2 are delay lines, G1 is a grating, P1-3 are polarizers, H1-3 are half waveplates, OPA1 and OPA2 are 5x5x5 mm KTP crystals cut for type II phase matching in the *xz* plane, BS1 and 2 are 50% reflecting mirrors, F1-3 are long pass filters, WL is the white light generation cell, M1 and M2 are dichroic mirrors, and L1-7 are lenses.

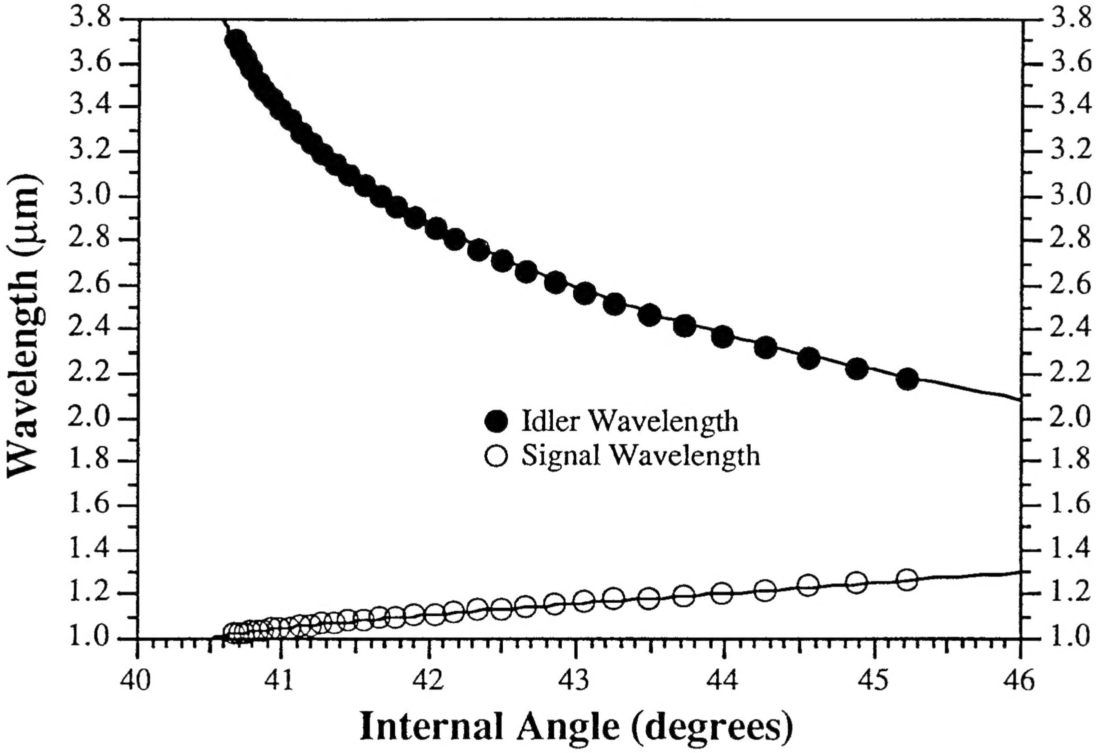

Figure 2

Experimental and theoretical tuning curves for type II (o-idler, e-signal, o-pump) phase matching in KTP. Empty circles are the experimental data for the signal beam and filled circles are the experimental data for the idler beam. Lines are the theoretical tuning curve from the Sellmeier equations (see text). The KTP crystals are 5x5x5 mm and cut at θ=42.65 and ϕ=0 degrees.

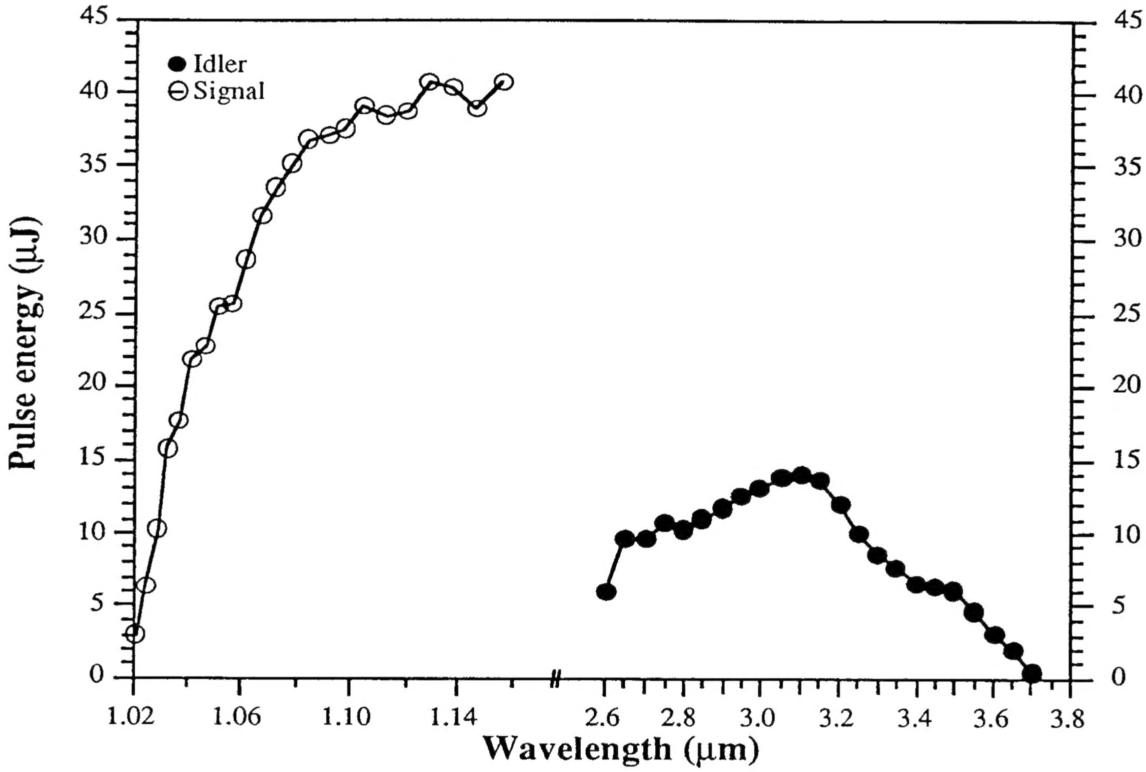

Figure 3

Output energy of signal, empty circles, and idler, filled circles, beams for the experimental tuning range in Figure 2. Measured energies were corrected for filter losses then plotted.